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Research Article

Utilization of the spent catalyst as a raw material for rechargeable battery production: The effect of leaching time, type, and concentration of organic acids

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Abstract. This study examines the potential use of the spent catalyst as a raw material for rechargeable batteries. The spent catalyst Ni/γ-Al_2O_3 still contains relatively high amounts of nickel. This indicates the potential use of the spent catalyst to be leached and purified for synthesizing nickel-based compounds so that it can be applied to rechargeable battery cathodes. In this study, the spent catalyst leaching process employed four types of organic acids: citric acid, lactic acid, oxalic acid, and acetic acid. The spent catalyst was leached under atmospheric conditions and room temperature. Organic acid concentrations were also varied at 0.1, 0.5, 1, and 2 M. The leaching process took place for 240 minutes, where sampling was conducted periodically at 30, 60, 120, 180, and 240 minutes. Experimental results showed that Ni (II) and Al (III) ions were successfully leached to the maximum when using 2M citric acids at a leaching time of 240 minutes. The conditions succeeded in leaching Ni (II) and Al (III) ions of 357.8 and 1,975.4 ppm, respectively. Organic acid, notably citric acid, has excellent potential for further development. Citric acid, as a solvent, has the ability to leach metal ions with high recovery. In addition, this acid is categorized as an eco-friendly and green solvent compared to inorganic acid. Thus, the leaching process can take place without harming the environment.

Keywords: Batteries, organic acid, spent catalyst, nickel, aluminium

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1. Introduction

The rapid development and production of electric vehicles worldwide have caused a rapid increase in rechargeable battery production. This has come about because electric vehicles require an energy storage system, like a rechargeable battery, to ensure that the vehicle can operate adequately (Maddu *et al.*, 2022; Thiruvonasundari & Deepa, 2021). This rechargeable battery comprises various components, such as an electrode (cathode and anode), electrolyte, and separator; those components are made of various compounds, for instance, metal alloys for the electrode, organic liquid for the electrolyte (Liang *et al.*, 2019; Cheng *et al.*, 2011). One of the critical elements in producing those batteries is nickel. In rechargeable batteries, nickel, in the form of an oxide, hydroxide, or other compounds, has a role as one of the materials that compose the cathode of rechargeable batteries (Ash *et al.*, 2020; Kiani *et al.*, 2010; Yan *et al.*, 2018; Zhang *et al.*, 2018). The choice of nickel as the cathode is due to the ability to provide higher storage capacity, higher energy density, and lower cost (Benayed *et al.*, 2021; Cui *et al.*, 2021; Y. Liu *et al.*, 2011; Xie *et al.*, 2019). Because of the importance of nickel in producing those batteries, it is necessary to pay attention to supplying nickel for the rechargeable battery industry so that there is no scarcity or shortage of nickel sources in the future.

Nickel can be obtained from primary and secondary mineral sources (Meshram *et al.*, 2018; Wanta *et al.*, 2020a). So far, the demand for nickel has been dominated by ore or concentrate, classified as primary mineral sources. However, a number of these major mineral sources will eventually be depleted, so the ore or concentrate will only be available temporarily. To maintain the sustainability of this nickel source, secondary mineral sources, such as spent catalysts or spent batteries, need to be utilized well (Garole *et al.*, 2020). The secondary mineral source is feasible because its metal components are still at high concentrations. For example, the nickel content in the spent catalyst amounts to 13–38% (Goel *et al.*, 2009; Sheik *et al.*, 2013; Wanta *et al.*, 2021). This value is higher than the nickel content in ore or concentrate. Thus, the effort of leaching nickel from the spent catalyst has excellent potential to be carried out, especially for nickel sources in producing these rechargeable batteries.

Nickel sources are one of many things to consider. The method of leaching nickel from those mineral sources also

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needs to be observed. Like the recovery of other metals, the leaching method is a proven method capable of leaching metal with nearly perfect effectiveness (Esmaeili *et al.*, 2020; Meshram *et al.*, 2016; Li *et al.*, 2015). So far, the application of this method has been dominated by using inorganic acid solvents such as sulphuric acid (Hosseini *et al.*, 2017; Ucyildiz & Girgin, 2017), hydrochloric acid (Le & Lee, 2010; Oediyani *et al.*, 2019), and nitric acid (Khalid & Athraa, 2017; Ramos-Cano *et al.*, 2016). However, this acid type has its drawbacks, primarily related to the environmental aspects involved (Astuti *et al.*, 2016). Demarco (2020) revealed that the use of inorganic acids in the metal leaching process allows the release of SO_3, Cl_2, and NO_x gases, which pose a risk to the environment. Furthermore, the acid waste generated after leaching is also harmful to the environment (Li *et al.*, 2010). As a result, greener solvents need to be applied so that the metal leaching process will not damage the environment in the future.

One of the green solvents that are feasible for application is organic acid one. Organic acids are classified as mild, generally biodegradable, and generate little or no environmental pollution (Pathak *et al.*, 2020). In addition, these organic acids can be produced using biotechnology, where the production of acids is carried out through fermentation pathways with the help of microorganisms (Angumeenal & Venkappayya, 2013; Chen & Nielsen, 2016). It is the forerunner of the emergence of a biological leaching process known as bioleaching (Srichandan *et al.*, 2019). Metallurgically, organic acid solvents are desirable and promising for application. The ability to remove metal ions is reasonably tested because, as an acid, this organic acid can still provide hydrogen ions which have a prominent role in the mechanism of the leaching process. In several studies that have been conducted, the performance and effectiveness of organic acids as solvents have been well-tested (Golmohammadzadeh *et al.*, 2018; Wanta *et al.*, 2022).

In the metal leaching process, Liu *et al.* (2021) have proven that organic acids can effectively leach various metals, namely Mn, Cd, Zn, and Pb, from plant combustion ashes. Their study also stated that citric acid was the best solvent for that particular process. Another study by Astuti *et al.* (2022) even explicitly proved that citric acid could operate better than inorganic acids in extracting lanthanum from the spent hydroprocessing catalyst. Citric acid is not the only type of organic acid that can be applied in the leaching process. Several similar studies yielded effective results in metal recovery from various secondary mineral sources when formic acid (Arslanoğlu & Yaraş, 2019), acetic acid (Behera & Parhi, 2016), and EDTA (Gaber, 2019) were applied as solvents.

This study focuses on the potential of the spent catalyst as a nickel source for rechargeable batteries. More specifically, leaching metal ions for this research study employs organic acid solvents. Many leaching studies like this have already been conducted. However, in the metallurgical context, differences in the characteristics of raw materials lead to different mechanisms and treatments for the leaching process. Each mineral resource has a uniqueness depending on its characteristics. A catalyst is an artificial mineral resource. It means that the catalyst comprises metal in the form of elements and compounds with a different mechanism to leach the metal ion. It is impossible for the metal in the form of elements to leach directly without additive compounds. Therefore, if metal elements dominate the catalyst composition, it will affect the mechanism of the leaching process. Thus, the potency of this spent catalyst needs to be studied further in terms of leaching the metal ion using organic acid.

This study emphasizes sustainable processes and green technology. The parameters studied are leaching time, acid type, and concentration. The operating condition of this process was conducted under operating conditions classified as a simple and safe process, such as atmospheric and low temperatures. The main objective of this research study is to observe the effect of leaching time, organic acid types (citric, oxalic, acetic, and lactic acids), and acid concentrations in recovering Ni (II) and Al (III) ions contained in the spent catalysts Ni/γ-Al_2O_3.

2. Material and Methods

2.1 Materials

The spent catalyst Ni/γ-Al_2O_3 employed as a source of metal ions in this study originated at PT. Petrokimia Gresik, Indonesia. This company is a fertilizer manufacturer; there is a reforming unit and applied Ni/γ-Al_2O_3 catalyst in that production. As a solvent, this leaching process applied four types of organic acids, namely citric acid (Merck), oxalic acid (Merck), acetic acid (Merck), and lactic acid (Merck). In addition, for analytical purposes, eriochrome cyanine R (ECR, Merck) and dimethylglyoxime (DMG, Merck) were used as complexing agents. All chemicals were dissolved using demineralized water.

The spent catalyst was analyzed using an x-ray fluorescence (XRF, Epsilon XLE PANalytical) instrument to determine the metal composition contained in the catalyst. The results of the analysis are presented in Table 1. This table shows that there are three elements, namely aluminium (Al), nickel (Ni), and calcium (Ca), which are the elements that dominate as well as compose this spent catalyst. These three elements in the catalyst are clearly visible in the mineral phases. Mineral phase testing was also conducted by analyzing the spent catalyst using an x-ray diffraction instrument (XRD, Bruker D8 Advance, Germany). The test results are presented in Fig. 1. Basically, this figure confirms the results of the XRF analysis and generates information about the mineral phase contained in the catalyst. The results of these characteristics still show that Al, Ni, and Ca dominate the mineral phase contained. Other components in Table 1 define various heavy metals contained in the spent catalyst, such as titanium (Ti), vanadium (V), zinc (Zn), gallium (Ga), strontium (Sr), yttrium (Y), zirconium (Zr), molybdenum (Mo), tellurium (Te), and iridium (Ir). The concentration of those elements is in part per million (ppm). This catalyst forms nickel in the pure nickel (Ni^o) and nickel oxide (NiO) crystalline phase. This leaching process is carried out without the use of additives that act as oxidizing agents. Thus, the nickel that can be leached only comes from the mineral NiO phase.

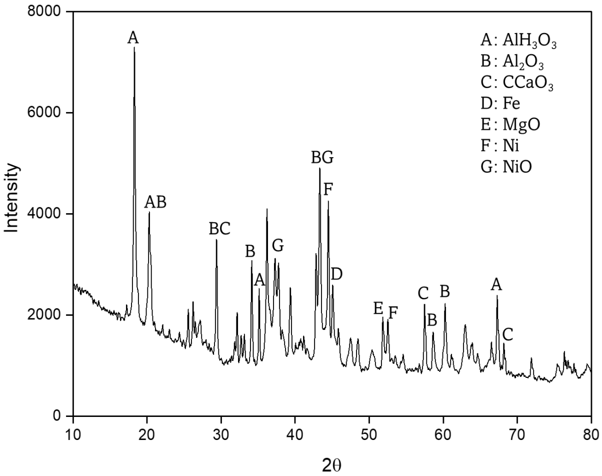

Fig. 1 XRD analysis results on the spent catalyst (raw material)

Table 1
XRF analysis results on the spent catalyst (raw material)

Component	Composition, %wt
Aluminium (Al)	38.2
Nickel (Ni)	37.7
Calcium (Ca)	22.6
Phosphorus (P)	0.4
Iron (Fe)	0.4
Potassium (K)	0.2
Silicon (Si)	0.2
Others	0.3

In the Ni^o phase, nickel will not be leached in this process because the characteristic of the phase is stable and cannot be dissolved in water. Nickel can only dissolve in water in the form of Ni (II) ions. This form will be produced from NiO compounds that react with hydrogen ions from acids. Meanwhile, for aluminium, the dominant form of aluminium that will react is aluminium oxide (Al_2O_3). This compound will react with hydrogen ions from acids and produce Al (III) ions, which dissolve in water.

2.2 Procedures

This leaching process was carried out using a set of equipment consisting of a 1 L glass bottle (as an extractor), a horizontal shaker incubator, and a sampler. First, the spent catalyst was milled and sieved to obtain the spent catalyst powder with a size of less than 149 microns. Seventy grams of spent catalyst powder were put into the extractor and mixed with 700 mL of the organic acid solution. After that, the extractor was conditioned on a shaker and was operated at 140 rpm. This leaching process was conducted at room temperature. The four types of organic acids were varied in this experiment. In addition, the concentration of each acid was varied and studied. For citric acid, acetic acid, and lactic acid, the studied concentration of those acid solutions was 0.1, 0.5, 1, and 2 M. Meanwhile, the concentration of the oxalic acid solution was only varied at 0.1, 0.5, and 1 M due to the maximum solubility of oxalic acid in water. By way of comparison, sulphuric acid was also used in this leaching process. This experiment was conducted to observe the potential of organic acid as a solvent compared to inorganic acid, which dominates the metal ions leaching process. In this experiment, the sulphuric acid's leaching process was designed at an acid concentration of 0.1 M for 240 minutes.

The sampling process for the metal ions analysis was carried out periodically at 30, 60, 120, 180, and 240 minutes. The sample was taken out from the extractor. The sample was separated between the solid and liquid phases using a centrifuge that operated at 6,000 rpm for 15 minutes. The formed supernatant (liquid phase) was taken to the amount of 10 mL and diluted to a solution volume of 20 mL. After that, the diluted sample was analyzed for the content of Ni (II) and Al (III) ions using a UV–vis spectrophotometer (Mapada UV–6100 PC). The analysis process was carried out using complexing agents, namely DMG for Ni (II) ions and ECR for Al (III) ions (Wanta *et al.*, 2020b).

2.3 Analysis of Ni (II) Ions

This analysis method was adapted from the article written by Haar & Westerveld (1948). Ten milliliters of sample solution were mixed with 1 mL of 2 N sulphuric acid solution, 2 mL of 20% sodium tartrate solution, 8 mL of 5% potassium persulphate solution, and 0.5 mL of 1% DMG solution. To this mixture was added 5 N of sodium hydroxide solution until the total volume of the mixture amounted to 50 mL. This sample was subsequently moved to a cuvette and was analyzed using a UV–vis spectrophotometer at a wavelength of 560 nm. The blank solution was produced by mixing 10 mL of demineralized water, a complexing agent, and 5 N sodium hydroxide until the total volume amounted to 50 mL.

2.4 Analysis of Al (III) Ions

This analysis method followed the procedure from the General Chemistry Laboratory, Eastern Michigan University (2018). Ten milliliters of sample solution were mixed with 2 mL of 0.02 N sulphuric acid solution, 20 mL of buffer solution, 2 mL of the ascorbic acid solution, and 10 mL of ECR solution. To this mixture was added demineralized water until the total volume amounted to 100 mL. This sample was subsequently moved to a cuvette and was analyzed using a UV–vis spectrophotometer at a wavelength of 535 nm. The blank solution was produced by mixing 10 mL of demineralized water and a complexing agent until the total volume amounted to 100 mL.

3. Results and Discussion

3.1 Effect of Leaching Time

Theoretically, the leaching time critically affects recovering metal ions in the spent catalyst. In this research study, the leaching time was varied from 0 to 240 minutes. The organic acid concentration was kept constant at 1 M. The experimental results on leaching time are presented in Fig. 2. This figure shows that the increase in Ni (II) ion recovery was accompanied by an increase in leaching time. The results show that from 30 to 240 minutes, the leaching process increased the recovery of Ni (II) ions between 2.41–3.91 times for the four types of acids. The profile of the experimental results that occur in the recovery of Ni (II) ions also occurs for the recovery of Al (III) ions. This can be observed in Fig. 3. The longer the leaching time, the bigger the opportunity that each molecule in the system remains in contact, especially the reactant molecules. As a result, the chemical reactions during the leaching process will react and produce more product molecules.

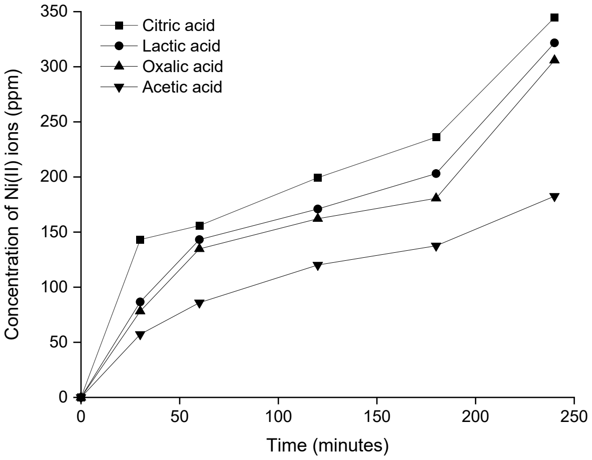

Fig. 2 Effect of leaching time on the Ni (II) ions recovery at an acid concentration of 1 M

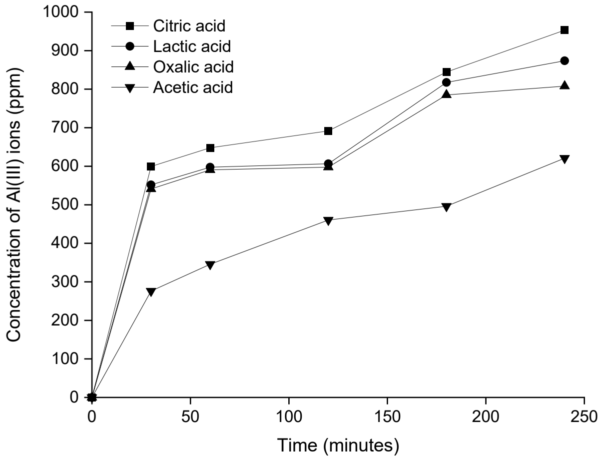

Fig. 3 Effect of leaching time on the Al (III) ions recovery at an acid concentration of 1 M

Time affects not only the step of chemical reactions but also molecular diffusion. The diffusion step experienced by each reactant and product molecule also takes time to move. The reactant molecules will diffuse from the bulk liquid to the surface of the liquid and the reaction site. On the other hand, the product molecules will move and diffuse from the reaction site to the liquid body. These two steps of diffusion will take place simultaneously so that in the pores/paths that tend to be narrow, all molecules will collide with each other. Thus, this diffusion step will require a long time to produce an optimal leaching process.

Fig. 2 and 3 provide an overview of the effect of time on metal ion recovery. These two figures explain how the behavior occurs during the leaching process. In the recovery of Al (III) ions (Fig. 3), there was a very significant increase in aluminium recovery in the first 30 minutes of the leaching process. After 30 minutes, the tendency to increase metal ion concentration occurred gradually. This phenomenon indicates that the leaching rate is swift at the beginning of the leaching time. This condition is related to the role of aluminium in catalyst production. Aluminium, in the form of γ-Al_2O_3 (γ-alumina), supports the catalyst (Trueba & Trasatti, 2005). It causes aluminium will occupy most of the area on the catalyst, especially on the surface of the catalyst powder. As a result, retrieving aluminium will be much easier and require faster leaching times, especially at the early stage of the leaching process. The opposite condition occurs in the uptake of Ni (II) ions. Fig. 2 shows that the concentration of Ni (II) ions in the bulk solution increases gradually. It proves that the recovery of Ni (II) ions takes longer. As the active site, nickel is scattered in a deeper position, so this recovery process takes longer, which is especially necessary for the diffusion process, both reactant molecules and product molecules.

In addition, the experimental results shown in Fig. 2 and 3 show the potential to obtain more and more metal ions. In this condition, it can be observed that there is no sign of equilibrium in the two graphs. This means that the maximum time used in this experiment differs from the time that gives optimal results. One of the main reasons this phenomenon occurs is that the temperature used in this research experiment is room temperature. Using this specific temperature will yield a slower process rate and result in a longer leaching time to obtain optimal leaching results. In this study, the main focus of learning is on room temperature. In the future, studies related to leaching temperature deserve to be considered and studied in greater depth so that its effect on the length of time for the leaching process can also be observed.

3.2 Effect of the Type of Organic Acid

In the leaching process, the choice of solvent is necessary. One type of solvent that can be used is an acid solution. Acid solutions are solvents that have been tested for leaching metal ions. It is due to hydrogen ions (H^+) which can be produced from the dissociation process when the acid is dissolved in water. This hydrogen ion will displace the metal ion from its anion, such as the oxide ion, so the metal ion can dissolve and form molecular compounds according to the acid's anion. The chemical reaction that occurs during the leaching process using organic acid is as follows (Behera & Mulaba-Bafubiandi, 2015; Golmohammadzadeh *et al.*, 2018; Simate *et al.*, 2010).

Organic acid dissociation step

Citric acid : $C_6H_8O_{7(aq)} \leftrightarrow (C_6H_5O_7)^{3-}_{(aq)} + 3H^+_{(aq)}$ (1)
Lactic acid : $C_3H_6O_{3(aq)} \leftrightarrow (C_3H_5O_3)^{3-}_{(aq)} + H^+_{(aq)}$ (2)
Oxalic acid : $C_2H_2O_{4(aq)} \leftrightarrow (C_2O_4)^{2-}_{(aq)} + 2H^+_{(aq)}$ (3)
Acetid acid : $C_2H_4O_{2(aq)} \leftrightarrow (C_2H_3O_2)^{-}_{(aq)} + H^+_{(aq)}$ (4)

Proton attack step

$$M_xO_{y(s)} + 2yH^+_{(aq)} \rightarrow xM^{2y/x+}_{(aq)} + yH_2O_{(l)} \quad (5)$$

where M stands for the metal element.

Complexation or chelation step

$$xM^{2y/x+}_{(aq)} + A^{z-}_{(aq)} \leftrightarrow M_zA_{2y/x(aq)} \quad (6)$$

where A is an anion from the dissociation step, for instance, citrate, lactate, oxalate, acetate ions.

This research study focuses on using organic acids as a solvent for metal ions in the spent catalyst. Four types of organic acids were studied for their effect on metal ion recovery: citric acid, lactic acid, oxalic acid, and acetic acid. The experimental results can be seen in Fig. 4, where the results are obtained when the acid concentration is 1 M and the extraction process takes place for 240 minutes. The experimental result shown in Fig. 4 provides information that the order of the types of organic acids from the best to the worst for leaching Ni (II) and Al (III) ions are citric acid – lactic acid – oxalic acid – acetic acid. Citric acid is the most suitable solvent due to the properties of the hydrogen ions. Equation (1) shows that hydrogen ions are produced in greater quantities than the other three organic acids.

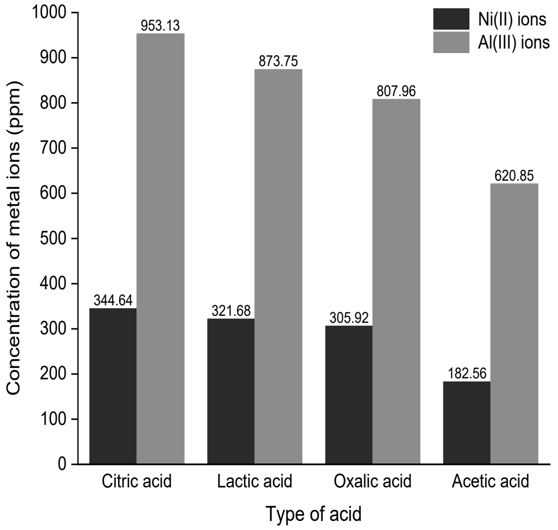

Fig. 4 Effect of leaching time on the metal ions recovery at an acid concentration of 1 M for 240 minutes

As a tricarboxylic acid, citric acid has the ability to donate three protons (H^+) per molecule when it dissociates in water. This means that citric acid also has three pKa values and can be observed in the following equation (Golmohammadzadeh *et al.*, 2018).

$$H_3C_6H_5O_{7(aq)} = H_2C_6H_5O_7^-{}_{(aq)} + H^+{}_{(aq)}\ (pKa_1 = 2.79) \quad (7)$$
$$H_2C_6H_5O_7^-{}_{(aq)} = HC_6H_5O_7^{2-}{}_{(aq)} + H^+{}_{(aq)}\ (pKa_2 = 4.30) \quad (8)$$
$$HC_6H_5O_7^{2-}{}_{(aq)} = C_6H_5O_7^{3-}{}_{(aq)} + H^+{}_{(aq)}\ (pKa_3 = 5.65) \quad (9)$$

The more hydrogen ions, the greater the probability of a proton attack. Thus, this organic acid can perform the metal leaching process better than other acids.

Instead of this, the pKa value of each acid can explain the phenomenon in the other three organic acids. The pKa values for lactic acid, oxalic acid, and acetic acid were 3.86, 4.19, and 4.76, respectively (Golmohammadzadeh *et al.*, 2018). The lower the pKa, the stronger the acidity. This indicates that the ability to donate protons (H^+ ions) in the system is also improving. This phenomenon explains why lactic acid can leach more Ni (II) and Al (III) ions compared to oxalic acid and acetic acid. However, the use of oxalic acid leads to another phenomenon where it also forms nickel oxalate precipitate. This phenomenon follows the following chemical reaction equation (Allen, 1953).

$$2NiSO_{4(aq)} + 2H_2C_2O_{4(aq)} \leftrightarrow [Ni(C_2O_4)_2]^- Ni^{2+}{}_{(aq)} + 2H_2SO_{4(aq)} \quad (10)$$

$$[Ni(C_2O_4)_2]^- Ni^{2+}{}_{(aq)} \rightarrow 2NiC_2O_4.2H_2O_{(s)} \quad (11)$$

As evidence of the formation of those compounds, testing of the residual solids resulting from leaching with 1 M oxalic acid solution was carried out to determine the mineral phase that occurred. The results of the analysis are presented in Fig. 5. The formation of nickel oxalate compounds during the leaching process is undesirable. Nickel oxalate compounds have very low solubility in water, so the nickel oxalate precipitation process will also coincide with the leaching process. Indeed, this phenomenon looks advantageous because it does not require precipitation or product crystallization at a later stage. However, this condition will be detrimental because the formed precipitate compound is mixed directly with the spent catalyst in the system. This will lead to new problems separating spent catalyst residues and nickel oxalate products. This separation process can be complicated or even classified as an impossible thing to accomplish. Thus, oxalic acid as a solvent in the metal leaching process is not recommended for use as a solvent.

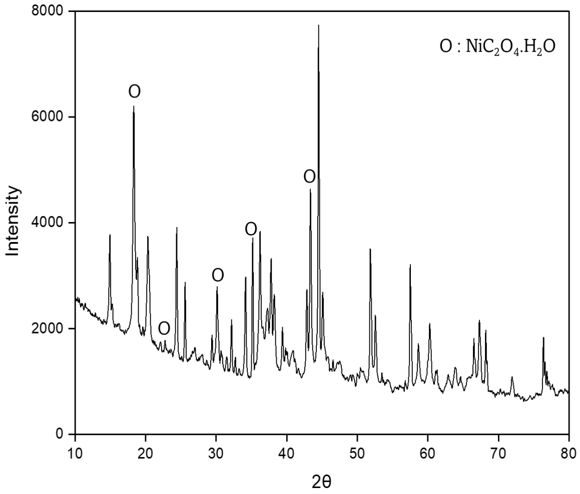

Fig. 5 XRD pattern on residue solid from leaching process using oxalic acid 1 M

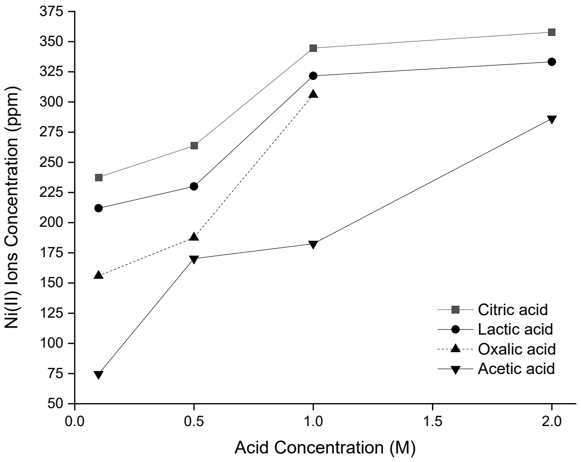

Fig. 6 Effect of acid concentration on the Ni (II) ions recovery for 240 minutes

3.3 Effect of Organic Acid Concentration

In studying the effect of organic acid concentrations, the utilization of citric acid, lactic acid, and acetic acid was carried out at concentrations of 0.1, 0.5, 1, and 2 M. Meanwhile, oxalic acid could not be carried out at concentrations of 2 M because of the solubility of that particular compound in water. The experimental results are presented in Fig. 6. These results showed similar results for the four types of organic acids. A higher concentration of acid will produce a more significant number of hydrogen ions. The hydrogen ions will attack the metal compound, for example, nickel and aluminium oxide, on the catalyst to produce water-soluble Ni (II) ions. Thus, a high acid concentration will increase the nickel's recovery in the solution. The phenomenon that occurs in Ni (II) ions is the same as in the leaching Al (III) ions. The experimental results for Al (III) ions can be observed in Fig. 7.

3.4 Organic Acid vs. Sulphuric Acid Employed for the Leaching Process

In this section, it between organic acid and sulphuric acid will be compared as a solvent for this leaching process. In hydrometallurgy, sulphuric acid is the most widely used type of solvent. The results of the comparison between the five types of acids are presented in Fig. 8, where the concentration used was 0.1 M. Sulphuric acid as a solvent gave much better leaching results than all the organic acids used in previous studies. As previously explained, the leaching process is strongly influenced by the strength of the acid, as seen from the number of hydrogen ions in the system. Sulphuric acid in water dissociates entirely according to the following chemical reaction equation (Vynnycky & Assunção, 2020).

$$H_2SO_{4(aq)} \rightarrow 2H^+{}_{(aq)} + SO_4^{2-}{}_{(aq)} \quad (12)$$

Compared with the four organic acids (equation 1–4), the hydrogen ion in sulphuric acid will ideally form according to stoichiometry. This is because the reaction is irreversible. On the other hand, in organic acids, the dissociation reaction is reversible, indicating that there is a balance factor in the formation of hydrogen ions. This results in citric acid, although it forms three hydrogen ions; in terms of the strength of sulphuric acid, which produces two hydrogen ions, it still has a higher level of acidity. As a result, in the leaching process with the same acid concentration, the performance of the sulphuric acid solvent is still superior.

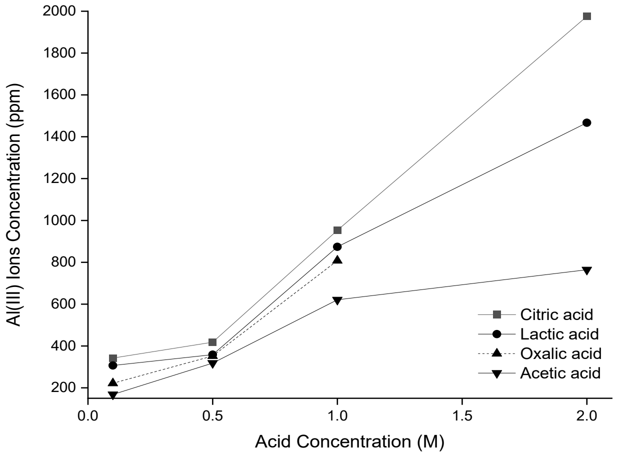

Fig. 7 Effect of acid concentration on the Al (III) ions recovery for 240 minutes

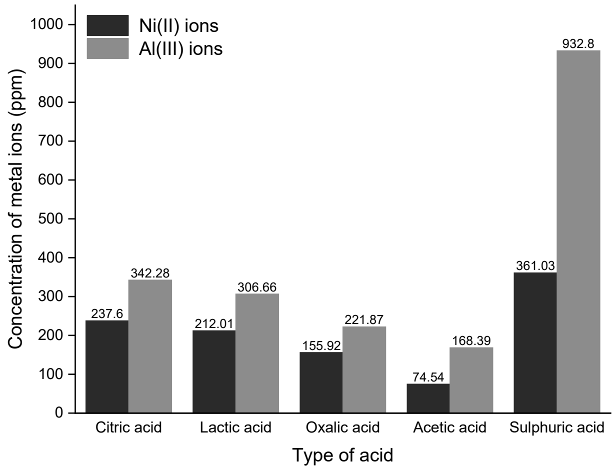

Fig. 8 The performance of organic and sulphuric acid in the leaching process of metal ions at an acid concentration of 0.1 M for 240 minutes

In addition, the use of organic acids (weak acids) raises other phenomena that will affect the mechanism of the metal leaching process. Equation (6) shows how complex compounds (chelates/ligands) are formed when metal ions bind to anions from organic acids. Meanwhile, this phenomenon does not occur using inorganic acids (strong acids) during the leaching process. The complex compound has a larger molecular size than the product molecules formed using inorganic acids (Wanta *et al.*, 2022). This condition will affect the mechanism and the leaching process's total rate. Large molecules will hinder the diffusion process of reactant and product molecules inside the solid. Thus, the rate of the leaching process with organic acids will be controlled by the diffusion step of the product molecules in the solid. This experiment needs to be designed by applying various kinetic models and will be the focus of further research in our future studies.

Even though sulphuric acid gives better results, using organic acids does not mean they do not have the potential and opportunities for further development. Moreover, this organic acid is an environmentally friendly solvent compared to sulphuric acid. Citric acid is a type of organic acid whose leaching behavior is similar to sulphuric acid. Astuti *et al.* (2016) conducted a comparative study between sulphuric acid and organic acids in the leaching process of nickel from Indonesian saprolite ore. In their study, the citric acid solvent was able to match the performance of sulphuric acid in their leaching process. This condition is based on the hope that organic acids, especially citric acid, deserve to be considered a suitable solvent for leaching metal ions. Indeed, several efforts need to be made to improve the performance of the leaching process using organic acids. The improvement effort in question can be in the form of increasing the operating temperature, reducing particle size, extending the operating time, and many other efforts.

4. Conclusion

Spent catalyst has excellent potential to be explored and utilized for its nickel resources as part of the raw material for manufacturing rechargeable battery cathodes. The metal from the catalyst can be leached through an extraction process using organic acid solvents. This experiment proves that citric acid, lactic acid, oxalic acid, and acetic acid can be applied as solvents. These four acids are believed to be green solvents compared to sulphuric acid, which is more widely used as a solvent. Based on the experimental results, citric acid is the solvent that gives the best leaching results compared to the other three organic acids. When the leaching process took place for 4 hours, the 2 M citric acid solution succeeded in leaching Ni (II) and Al (III) ions of 357.8 and 1,975.4 ppm, respectively. The recovery of metal ions with citric acid results are still lower than the ones with sulphuric acid as a solvent. However, citric acid can still be developed further to have the same or better performance than sulphuric acid. Various alternatives for manipulating the operating parameters of the leaching process, such as temperature, particle size, the addition of additives, and others, can be conducted to increase the process rate. Thus, the extraction of metal ions from the spent catalyst is suitable for further study to obtain maximum results and meet the demand for metal ions as a raw material for producing rechargeable batteries. The results obtained from this research study are expected to form a solution in supplying the need for nickel as a raw material for producing rechargeable batteries.

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